Spin Heat Accumulation Induced by Tunneling from a Ferromagnet

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An electric current from a ferromagnet into a nonmagnetic material can induce a spin-dependent electron temperature. Here, it is shown that this spin heat accumulation, when created by tunneling from a ferromagnet, produces a non-negligible voltage signal that is comparable to that due to the coexisting electrical spin accumulation and can give a different Hanle spin precession signature. The effect is governed by the spin polarization of the Peltier coefficient of the tunnel contact, its Seebeck coefficient, and the spin heat resistance of the nonmagnetic material, which is related to the electrical spin resistance by a spin-Wiedemann-Franz law. Moreover, spin heat injection is subject to a heat conductivity mismatch that is overcome if the tunnel interface has a sufficiently large resistance.

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Creation and detection of spin information are at the heart of spintronics, the study and use of spin degrees of freedom [1]. Electronic spin transport is described by a two-channel model where transport is separately considered for each spin ($\sigma = \uparrow, \downarrow$) population [2–4]. When a current is applied between a ferromagnetic contact and a nonmagnetic material, it induces a spin accumulation $\Delta\mu = \mu^{\uparrow} - \mu^{\downarrow}$ described by a splitting of the electrochemical potentials μ^{σ} of the two spin channels in the nonmagnetic material [5–8]. Direct electrical detection of the spin accumulation is achieved via the Hanle effect, where a magnetic field induces spin precession and suppresses $\Delta\mu$, giving a measurable voltage signal. Interestingly, spin current in ferromagnetic tunnel contacts can be created both by an electrical bias [9,10] or by a thermal bias [11,12]. The latter approach of thermal spin injection is possible due to the spin dependence of thermoelectric properties in magnetic materials and nanodevices, which lead to interactions between spin and heat transport currently studied in the field of spin caloritronics [13,14]. This raises the question: do Hanle measurements only detect a difference in electrochemical potentials μ^{σ}, as hitherto assumed, or also a difference in temperatures T^{σ} between the two spin channels?

In this Letter, we address the creation and detection of a spin heat accumulation $\Delta T_s = T^{\uparrow} - T^{\downarrow}$ in a nonmagnetic material via a ferromagnetic tunnel contact. It is considered, here, that tunneling transport is accompanied by a spin-dependent heat flow if the Peltier coefficient of the tunnel contact depends on spin, and that this produces a spin heat accumulation and an additional contribution to the voltage signal in a Hanle measurement. Spin heat accumulation is a concept previously studied theoretically within the context of metallic spin-valve structures [15–17], and, only very recently, it has been observed as a spin-dependent heat conductance in metallic current-perpendicular-to-plane spin-valve nanopillars [18]. Here, we provide an explicit evaluation for the spin heat accumulation at the tunnel interface between a ferromagnet and a nonmagnetic material. Notably, we introduce the notion of an associated heat conductivity mismatch, similar to that for spin accumulation [19–23], which limits the magnitude of spin heat accumulation and can be overcome with the tunnel interface. Most importantly, we show that the widely employed Hanle measurement to detect spin accumulation has another contribution from the spin heat accumulation that can be comparable in magnitude and has a line width set by the spin heat relaxation time. It cannot be neglected *a priori* and needs to be considered for a correct interpretation of experimental data.

We consider the case of a three-terminal geometry, where the same contact is used for driving an electrical current and measuring the voltage signal, as commonly used for spin injection into semiconductors [7,8], although the basic physics also applies to other device geometries, such as the nonlocal one. Such a tunnel junction with a ferromagnetic electrode and a nonmagnetic semiconductor electrode is depicted in Fig. 1. We describe each spin population in the nonmagnetic material by a Fermi-Dirac distribution with spin-dependent temperatures $T^{\uparrow}$ and $T^{\downarrow}$. This is strictly valid only when thermalization within each spin channel is sufficiently fast compared to energy exchange between the spin channels. In general, the distributions could be nonthermal and we should regard T^{σ} as effective temperatures [17,18]. For the ferromagnet we assume negligible spin and spin heat accumulations due to stronger spin-flip and inelastic scattering processes, so both spin channels are equilibrated at T_F. We define an average electron

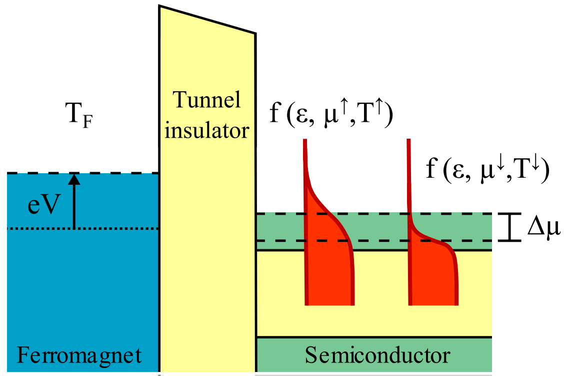

FIG. 1 (color online). Energy band diagram of a ferromagnet-insulator-semiconductor tunnel junction. The electrons in the semiconductor have spin-dependent temperatures $T^\uparrow$ and $T^\downarrow$, whereas those in the ferromagnet are at T_F. Also, a spin accumulation exists in the semiconductor, described by a spin splitting $\Delta\mu = \mu^\uparrow - \mu^\downarrow$ of the electrochemical potential. The distribution functions are indicated by the red lines.

temperature $T_0 = (T^\uparrow + T^\downarrow)/2$ in the nonmagnetic material, and a temperature difference $\Delta T_0 = T_0 - T_F$ across the contact. The charge tunnel currents I^σ and the electronic heat currents $I^{Q,\sigma}$ for each spin channel are then given by [12]

$$I^\sigma = G^\sigma\left(V \mp \frac{\Delta\mu}{2e}\right) + L^\sigma\left(\Delta T_0 \pm \frac{\Delta T_s}{2}\right), \qquad (1)$$

$$I^{Q,\sigma} = -\kappa^{\mathrm{el},\sigma}\left(\Delta T_0 \pm \frac{\Delta T_s}{2}\right) + G^\sigma S^\sigma T_0\left(V \mp \frac{\Delta\mu}{2e}\right), \qquad (2)$$

with V the voltage across the junction and $\kappa^{\mathrm{el},\sigma}$ the electronic heat conductance of the tunnel barrier in units of [W m^{-2} K^{-1}]. Charge currents I^σ are in units of [A m^{-2}], conductances G^σ in [Ω^{-1} m^{-2}] and heat currents $I^{Q,\sigma}$ in [W m^{-2}]. By definition, $I > 0$ and $I^Q > 0$ correspond to electron flow and heat flow, respectively, from the ferromagnet to the semiconductor. Furthermore, the Onsager coefficient L (thermoelectric conductance) is positive when the conductance below the Fermi energy is larger than that above it, so the spin-dependent Seebeck coefficient $S^\sigma = -L^\sigma/G^\sigma < 0$ for holelike transport [24].

First, we proceed to find the spin current and the spin heat current injected into the nonmagnetic material. The spin current $I_s = I^\uparrow - I^\downarrow$, and the charge current $I = I^\uparrow + I^\downarrow$, are obtained from Eq. (1)

$$I = GV - P_G G\left(\frac{\Delta\mu}{2e}\right) + L\Delta T_0 + P_L L\left(\frac{\Delta T_s}{2}\right), \qquad (3)$$

$$I_s = P_G GV - G\left(\frac{\Delta\mu}{2e}\right) + P_L L\Delta T_0 + L\left(\frac{\Delta T_s}{2}\right), \qquad (4)$$

where we have defined the total conductances $G = G^\uparrow + G^\downarrow$ and $L = L^\uparrow + L^\downarrow$, and their spin polarizations $P_G = (G^\uparrow - G^\downarrow)/(G^\uparrow + G^\downarrow)$ and $P_L = (L^\uparrow - L^\downarrow)/(L^\uparrow + L^\downarrow)$.

The total heat current $I^Q = I^{Q,\uparrow} + I^{Q,\downarrow} + I^{Q,\mathrm{ph}}$ contains, in addition to the heat flow by electrons, a dominant contribution due to phonon transport across the tunnel contact. It is given by $I^{Q,\mathrm{ph}} = -\kappa^{\mathrm{ph}}(T^{\mathrm{ph}} - T_F)$, where κ^{ph} is the phonon heat conductance of the barrier (usually dominated by the interfaces [25]), T^{ph} is the phonon temperature in the nonmagnetic electrode, and we assume that, in the ferromagnet, phonons and electrons are fully equilibrated at T_F. Phonon heat flow is not parametrized by the spin variable and does not contribute to the spin heat current across the barrier. Thus, the injected spin heat current $I_s^Q = I^{Q,\uparrow} - I^{Q,\downarrow}$ is only due to the electrons and can be obtained from Eq. (2)

$$I_s^Q = -P_\kappa^{\mathrm{el}}\kappa^{\mathrm{el}}\Delta T_0 - \kappa^{\mathrm{el}}\left(\frac{\Delta T_s}{2}\right) - P_L L T_0 V + L T_0\left(\frac{\Delta\mu}{2e}\right), \qquad (5)$$

where we have defined the total electronic heat conductance of the tunnel contact $\kappa^{\mathrm{el}} = \kappa^{\mathrm{el},\uparrow} + \kappa^{\mathrm{el},\downarrow}$, its polarization $P_\kappa^{\mathrm{el}} = (\kappa^{\mathrm{el},\uparrow} - \kappa^{\mathrm{el},\downarrow})/(\kappa^{\mathrm{el},\uparrow} + \kappa^{\mathrm{el},\downarrow})$ and used the relation $S^\sigma G^\sigma = -L^\sigma$.

Now, we can evaluate the contribution of the spin heat accumulation to a Hanle measurement. Typically, a Hanle measurement involves the application of a constant electrical current I at the tunnel junction while spin precession in a magnetic field B perpendicular to the injected spins causes $\Delta\mu$ to go to zero. The decrease in $\Delta\mu$ depends on the product of the spin-relaxation time τ_s and the Larmor frequency $\omega_L = g\mu_B B/\hbar$, with g the Landé g factor, μ_B the Bohr magneton, and $\hbar$ the reduced Planck constant. Importantly, spin precession would also cause ΔT_s to go to zero. This can be understood by considering a packet of hot spins polarized along $+x$ and an equal amount of cold spins polarized along $-x$. A perpendicular field along z causes a precession of each spin in the x–y plane and, thereby, a periodic oscillation of the temperature of the electrons with spin pointing along $+x$ (or $-x$) [26]. If the precession frequency ω_L is much larger that the inverse of the time constant τ_Q, associated with relaxation of the spin heat accumulation, then the time-average of ΔT_s goes to zero. Therefore, for such an electrically driven junction, and assuming that the spin-averaged temperatures of the electrodes remain constant, the corresponding Hanle signal $\Delta V_{\mathrm{Hanle}} = V - V|_{\Delta\mu,\Delta T_s\to 0}$ can be obtained from Eq. (3)

$$\Delta V_{\mathrm{Hanle}} = \left(\frac{P_G}{2e}\right)\Delta\mu + \left(\frac{P_L S}{2}\right)\Delta T_s. \qquad (6)$$

In addition to the well-known Hanle signal arising from the spin accumulation $\Delta\mu$, there is a second, up to now neglected, contribution due to the spin heat accumulation ΔT_s. Note that the Hanle curve $\Delta V_{\mathrm{Hanle}}$ vs B, due to suppression of $\Delta\mu$, is Lorentzian [1] and has a width that is inversely proportional to the spin-relaxation time τ_s.

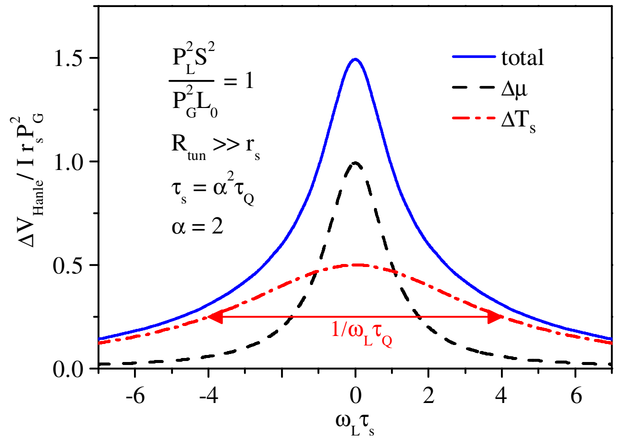

FIG. 2 (color online). Hanle signal in a three-terminal configuration and the contributions corresponding to spin accumulation $\Delta\mu$ and spin heat accumulation ΔT_s. The relative magnitude of the signals is obtained from Eq. (14), assuming $(P_L S)^2/(P_G^2 L_0) = 1$, $\alpha = 2$, and $R_{\text{tun}} \gg r_s$. We consider a spin heat relaxation time $\tau_Q < \tau_s$, given by $\tau_s/\tau_Q = (\lambda_s/\lambda_Q)^2 = \alpha^2$.

Similarly, we expect that the suppression of ΔT_s yields a Lorentzian Hanle curve having a width that is inversely proportional to the spin heat relaxation time τ_Q, such that $\Delta T_s(B) \propto [1 + (\omega_L \tau_Q)^2]^{-1}$. If τ_Q is sufficiently different than τ_s, then the total Hanle signal will consist of two superimposed Hanle curves with different widths, as depicted in Fig. 2. We remark that the latter directly follows from the sum of two independent contributions to the voltage, conform to Eq. (6). Interestingly, if a spin heat accumulation is present, interpreting the total Hanle signal purely in terms of a spin accumulation would lead to an underestimation of the spin-relaxation time τ_s.

Next, we evaluate the created spin accumulation $\Delta\mu$ and spin heat accumulation ΔT_s in the nonmagnetic material. We consider a steady-state condition in which the spin current I_s injected by tunneling is balanced by the spin current due to spin relaxation processes in the material [5], occurring over the spatial extent of $\Delta\mu$. Similarly, the spin heat current I_s^Q injected by tunneling is balanced by the heat current between the two spin populations due to spin relaxation and inelastic scattering processes [17], occurring over the spatial extent of ΔT_s. To relate accumulations and injected currents, we define a spin resistance r_s and a spin heat resistance r_s^Q of the nonmagnetic material

$$\Delta\mu = 2eI_s r_s, \tag{7}$$

$$\Delta T_s = 2I_s^Q r_s^Q, \tag{8}$$

where r_s is a phenomenological parameter that describes the conversion of the spin current I_s injected by tunneling, into a spin accumulation with a value of $\Delta\mu$ right at the tunnel interface, as before [27]. This definition does not require us to assume any specific profile of the spin accumulation in the nonmagnetic material. If we do assume that the spin accumulation decays exponentially away from the tunnel interface with a spin-relaxation length λ_s [5], then the spin resistance per unit area is $r_s = \rho\lambda_s$, where ρ is the resistivity of the nonmagnetic material [21,28,29]. Similarly, the parameter r_s^Q is defined in terms of the injected spin heat current I_s^Q and the spin heat accumulation ΔT_s right at the tunnel interface.

Using the definition above for spin heat resistance, we can obtain ΔT_s from Eqs. (5) and (8)

$$\Delta T_s = \left\{\frac{r_s^Q R_{\text{tun}}^{Q,\text{el}}}{R_{\text{tun}}^{Q,\text{el}} + r_s^Q}\right\} \times \left[\frac{(2P_L V - \Delta\mu/e)}{R_{\text{tun}}} ST_0 - \frac{2P_\kappa^{\text{el}}}{R_{\text{tun}}^{Q,\text{el}}}\Delta T_0\right], \tag{9}$$

where $R_{\text{tun}} = 1/G$ is the tunnel resistance, and $R_{\text{tun}}^{Q,\text{el}} = 1/\kappa^{\text{el}}$ is the electronic thermal resistance of the tunnel barrier. The two terms within the square brackets represent the sources of spin-dependent heat flow. The first one is due to the spin heat current that accompanies the charge current across the tunnel contact, governed by the spin polarization P_L of the Peltier coefficient [30]. The second term is present when there is a temperature bias across the junction, driving a spin-dependent heat flow if the heat conductance of the tunnel barrier is spin dependent ($P_\kappa^{\text{el}} \neq 0$) [18]. If transport through the tunnel barrier is elastic, the tunnel resistance and the electronic thermal resistance of the tunnel barrier are interrelated by the Wiedemann-Franz law [31]

$$R_{\text{tun}}^{Q,\text{el}} = \frac{1}{\kappa^{\text{el}}} = \frac{1}{L_0 T_0 G} = \frac{R_{\text{tun}}}{L_0 T_0}, \tag{10}$$

with $L_0 \approx 2.45 \times 10^{-8}\ \text{V}^2\,\text{K}^{-2}$ the Lorentz number. This allows us to estimate the magnitude of $R_{\text{tun}}^{Q,\text{el}}$ and, together with Eqs. (3) and (9), to obtain an explicit evaluation of the spin heat accumulation ΔT_s in terms of the driving current I. Furthermore, we can also obtain an explicit evaluation for the spin accumulation $\Delta\mu$ from Eqs. (3), (4), and (7). The resulting expressions are

$$\Delta\mu = \left\{\frac{r_s R_{\text{tun}}}{R_{\text{tun}} + (1 - P_G^2) r_s}\right\}\left(\frac{e}{R_{\text{tun}}}\right)[2P_G R_{\text{tun}} I + (1 - P_G^2)(S^\uparrow - S^\downarrow)\Delta T_0 - (1 - P_L P_G) S \Delta T_s], \tag{11}$$

$$\Delta T_s = \left\{\frac{r_s^Q R_{\text{tun}}^{Q,\text{el}}}{R_{\text{tun}}^{Q,\text{el}} + (1 - \frac{P_L^2 S^2}{L_0}) r_s^Q}\right\}\left(\frac{ST_0}{R_{\text{tun}}}\right)\left[2P_L R_{\text{tun}} I - (1 - P_L P_G)\frac{\Delta\mu}{e} + 2\left(P_L - P_\kappa^{\text{el}}\frac{L_0}{S^2}\right) S \Delta T_0\right]. \tag{12}$$

Note the similarity among the terms between curly brackets in Eqs. (11) and (12). The term in Eq. (11) corresponds to the known issue of conductivity mismatch [19–23] which limits the magnitude of the spin accumulation due to back flow of the spins into the ferromagnet when $R_{\mathrm{tun}} < r_s$. Remarkably, the term in Eq. (12) alludes to an analogous notion of a heat conductivity mismatch: if the heat resistance $R_{\mathrm{tun}}^{Q,\mathrm{el}}$ of the tunnel barrier is smaller than the spin heat resistance r_s^Q of the nonmagnetic material, then the spin heat accumulation is reduced by back flow of the spin heat into the ferromagnet. In order to overcome the heat conductivity mismatch, one needs to fulfill the condition $R_{\mathrm{tun}}^{Q,\mathrm{el}} \gg r_s^Q$. Note that this is governed by the electronic heat resistance of the tunnel contact, since phonons cannot transport spin heat across the tunnel barrier. This concept is crucial to the creation of a large ΔT_s, which in recent experimental work was limited by highly transparent metallic contacts [18].

Finally, we evaluate the magnitude of the spin heat accumulation and its corresponding contribution to the Hanle signal. Both are fully described by Eqs. (6), (11), and (12). In the following, we avoid the, in general, lengthy solutions and proceed to make a few practical simplifications. First, we consider a junction that is driven electrically, not thermally, so that the spin accumulation is dominated by electrical spin injection. We, therefore, neglect the second term (due to Seebeck spin tunneling [11]) and third term (due to a nonzero spin heat accumulation ΔT_s) in Eq. (11), and retain only the first term proportional to I. Note that the last term $S\Delta T_s$ is expected to be smaller than 1 mV, and, thus, small compared to $R_{\mathrm{tun}}I$. Similarly, the dominant term in Eq. (12) for ΔT_s is the first term, proportional to I (due to the spin-dependent Peltier effect). This is valid since $\Delta\mu/e \ll R_{\mathrm{tun}}I$, and ΔT_0 is typically smaller than $SR_{\mathrm{tun}}I/L_0 \approx 40\mathrm{K}$ for reasonable values of $S = 100\ \mu\mathrm{V\,K}^{-1}$ and $R_{\mathrm{tun}}I = 100$ mV.

Still, there is one parameter with an unknown magnitude, the spin heat resistance of the nonmagnetic material r_s^Q. It can be expressed as [26]

$$r_s^Q = \frac{r_s}{L_0 T_0}\frac{1}{\alpha}. \tag{13}$$

This constitutes a type of spin-Wiedemann-Franz law, relating the electronic spin resistance to the spin heat resistance. The parameter α takes into account the inelastic scattering processes occurring within the nonmagnetic material which increase the interspin heat exchange. These microscopic processes, described in Ref. [17], correspond to electron-electron and electron-phonon interactions that cause relaxation of the spin heat accumulation and decrease τ_Q. We remark that α is related to the concept of a spin heat relaxation length $\lambda_Q = \sqrt{D\tau_Q}$ in the regime of diffusive (heat) transport, with the corresponding diffusion constant D being the same as that for charge transport since electronic heat transport is associated with electrons only. In the case of an exponentially decaying spin (heat) accumulation, it follows that $\alpha = \lambda_s/\lambda_Q$, which, in recent work [18], has been estimated to be $\lambda_s/\lambda_Q \approx 5$ for Cu at room temperature. Therefore, α can indeed be of order one. In general, we may expect $\alpha > 1$, although the definition [26] of α does not preclude a value smaller than unity. At low temperatures, inelastic scattering processes are reduced [17], so we expect elastic spin-flip scattering to be the dominant spin relaxation mechanism and $\alpha \to 1$. Using this result, we finally obtain for the Hanle signal

$$\frac{\Delta V_{\mathrm{Hanle}}}{I} = r_s (P_G)^2 \left\{\frac{R_{\mathrm{tun}}}{R_{\mathrm{tun}} + (1-P_G^2) r_s}\right\} + \frac{r_s}{\alpha}\left(\frac{P_L^2 S^2}{L_0}\right)\left\{\frac{R_{\mathrm{tun}}}{R_{\mathrm{tun}} + (1-\frac{P_L^2 S^2}{L_0})\frac{r_s}{\alpha}}\right\}, \tag{14}$$

where the first term is due to $\Delta\mu$ and the second term is due to ΔT_s.

The relative magnitude of the two contributions to the Hanle signal is governed by the ratio P_L/P_G, by α, and by the factor S^2/L_0. The latter is unity for $S \simeq 157\ \mu\mathrm{V/K}$, which, in the Sommerfeld approximation, is the maximum value with a non-negative entropy production, as required by the second law of thermodynamics [32]. Previous work has shown that the Seebeck coefficient of a tunnel junction is indeed in the order of 100 μV/K [11,33–35], and for the case of ferromagnetic electrodes is enhanced by magnons [36,37]. If $P_L \sim P_G$, the Hanle signals due to ΔT_s and $\Delta\mu$ are, then, comparable in magnitude. Interestingly, both contributions always show the same sign, because there are only quadratic terms.

We conclude that the spin heat accumulation can make a significant contribution to the Hanle signal that cannot be neglected *a priori*. In principle, it can even be larger than the regular Hanle signal from the spin accumulation if $P_L > P_G$, which can occur when $S^\uparrow \neq S^\downarrow$ since we have $P_L = P_G - (1-P_G^2)(S^\uparrow - S^\downarrow)/(2S)$. However, the resulting enhancement of the Hanle signal is not sufficiently large to explain recent experiments in which Hanle signals that scale with the tunnel resistance and are many orders of magnitude larger than predicted by theory are observed [38–44], as recently reviewed [27,45]. It would require $\alpha \ll 1$ by several orders of magnitude. To estimate the magnitude of the spin heat accumulation, we use Eq. (12) in the regime without heat conductivity mismatch ($R_{\mathrm{tun}}^Q \gg r_s^Q$), and retain only the leading term, so we obtain $\Delta T_s = 2 r_s S P_L I/L_0\alpha$. For reasonable parameters at room temperature of $r_s = 25\ \Omega\,\mu\mathrm{m}^2$, $S = 100\ \mu\mathrm{V/K}$, and $P_L = 50\%$, a modest current density of $10^7\ \mathrm{A\,m}^{-2}$ would yield a spin heat accumulation of $\Delta T_s \approx 1$ K (for $\alpha = 1$).

The creation, manipulation, and detection of spin heat accumulation is a subject that is still in its infancy. The realization that it contributes to Hanle measurements, given the spin dependence of Peltier coefficients in magnetic tunnel contacts, makes it a non-negligible factor that needs to be taken into account in current studies of spin injection. It affects the magnitude and width of the Hanle curve and its variation with temperature, and is also expected to be present in (non)local spin-valve measurements. The analogy between spin and heat transport, here made explicit by the concept of a heat conductivity mismatch, opens opportunities to address fundamental questions about the relaxation of a spin heat accumulation. How do inelastic processes affect the magnitude of the parameter α in the spin-Wiedemann-Franz law and the width of the Hanle curve, and how is this behavior modified under the presence of a strong spin-orbit coupling? And is it possible that $\alpha < 1$, meaning that the spin heat relaxes slower than the spin accumulation?

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